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A Summary of Information in the Literature

UNITED STATES DEPARTMENT OF THE INTERIOR

Information Circular 8756

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By Ann G. Kim

UNITED STATES DEPARTMENT OF THE INTERIOR
Cecil D. Andrus, Secretary

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CONTENTS

GLOSSARY

Adiabatic--system in which there is no exchange of heat with the surroundings.

Air flow rate--the volume of air to which the coal is exposed in unit time.

Calorimetry--system for measuring heat produced by a chemical reaction.

Combustion--a chemical process in which heat is released or "evolved." Common usage of this term suggests open flame, but its technical application relates only to the release of energy, which may or may not result in open flame.

Endothermic--requiring or absorbing heat.

Exothermic--releasing or evolving heat.

Heat of oxidation--amount of energy released by reaction of coal with oxygen.

Heat of wetting--amount of energy released by adhesion of water to the coal surface.

Heat sink--large reservoir held at a constant temperature, in which the relatively small amount of heat generated by a reaction is dissipated, causing no change in the temperature of the overall system.

Index of combustibility--objective indicator of the tendency toward spontaneous heating.

Isothermal--system in which there is no change in temperature.

Moisture content--amount of water adsorbed on the surface of the coal.

Oxidation--reaction with oxygen.

Rank--degree of metamorphism; defined by fixed carbon content and heating value.

Spontaneous heating--heating that occurs without an external ignition source; also called "self-heating" or "spontaneous combustion." Energy released by this process may be sufficient to cause a fire.

Wetting--adhesion of a liquid to a surface.

LABORATORY STUDIES ON SPONTANEOUS HEATING OF COAL

A Summary of Information in the Literature

by

Ann G. Kim[1]

ABSTRACT

This Bureau of Mines report summarizes the methods and results of many of the laboratory studies on spontaneous heating of coal published in the last 50 years. It includes a brief description of experimental methods, indices of combustibility, and important contributing factors. Adiabatic calorimetry, isothermal calorimetry, O_2 sorption systems, or temperature differential are the most frequently used experimental methods. Indices of combustibility include various heating rates, crossing point temperature, ignition temperature, and CO index. Factors that affect the spontaneous heating of coal include air flow rate, changes in moisture content, particle size, rank, temperature, pyrite content, geological factors, and mining practice. Experimental conditions and results from 20 papers are summarized in an appendix, and the bibliography contains 45 references.

INTRODUCTION

The expected decrease in the availability of premium fuels has focused attention on the resource potential of low-rank coals (subbituminous coals and lignites), which have a heating value of less than 13,000 Btu. There are an estimated 485 billion tons of subbituminous coal and 478 billion tons of lignite in the United States, mostly in the continental States west of the Mississippi (1).[2] Increased production and utilization of low-rank western coals present many problems, both technical and economic. One serious problem associated with mining, transportation, and storage of low-rank coals is spontaneous heating.[3] Fires due to spontaneous heating of coal may occur in the high wall of surface mines, on trains, or in storage piles, and present a potentially fatal hazard in underground mines. Spontaneous heating, which is a common problem with some European and Soviet coals, has been the subject of

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[1] Chemist, Pittsburgh Mining and Safety Research Center, Pittsburgh, Pa.
[2] Underlined numbers in parentheses refer to items in the bibliography preceding the appendix.
[3] The terms "spontaneous heating," "self-heating," and "spontaneous combustion" are commonly used to describe the same process. Although "spontaneous heating" is used in this report, the terms are technically equivalent and are used interchangeably in the literature.

research for over 100 years. The extensive literature on this subject can be divided into four general categories: Experimental studies, prevention, detection, and control. The experimental studies compose the largest segment and serve as the theoretical basis for the other topics. These studies have demonstrated that the most important factor in spontaneous heating is the susceptibility of the coal, with low-rank coals more susceptible than high-rank coals. Additional factors that affect the rate at which spontaneous heating will occur include changes in the moisture content of the coal, air flow rate, particle size, temperature, pyrite content, geological factors, and mining practice. Based on the information available from many experimental studies, it might be expected that problems due to spontaneous heating would be subject to a well-defined program of prevention, detection, and control. Unfortunately, this is not the case. At present, there is no simple, universally applicable test for combustibility, no generally accepted index of combustibility, and no simple effective method of preventing spontaneous heating. Prevention is based on suppressing factors that favor spontaneous heating, such as accumulation of fine coal particles, inefficient heat dissipation, and differences in moisture content of the air and the coal. Control of spontaneous heating is usually based on previous experience, and includes mining practice to reduce risk and detection of incipient combustion before a fire occurs.

Because initial steps in evaluating the potential danger from spontaneous heating involve determining a coal's susceptibility to it, and defining the factors that increase the hazard, information from many experimental studies on these topics is summarized in this Bureau of Mines report. It includes (1) a brief discussion of factors affecting spontaneous heating, indices of combustibility, and experimental methods; (2) a bibliography of 45 pertinent references; and (3) an appendix containing a tabular comparison of the experimental conditions and results of 20 important papers.

FACTORS AFFECTING SPONTANEOUS HEATING OF COAL

Several factors have a significant effect on the rate of spontaneous heating[4] of coal in the laboratory and in the mine.

Air Flow Rate.--The air flow rate is a complex factor because air both provides oxygen for oxidation of the coal and dissipates the heat generated by oxidation. A very high flow rate provides almost unlimited oxygen, but dissipates heat efficiently. A low flow rate restricts the amount of oxygen available, but allows heat generated to remain in the coal. In mining, a critical flow rate would be one that provides sufficient oxygen for widespread oxidation but does not dissipate the heat generated.

Particle size.--Particle size has an inverse relationship to spontaneous heating of coal. The smaller the coal particle, the greater is the exposed surface area and the greater is the tendency toward spontaneous heating. In laboratory experiments, smaller particle size fractions are used in order to obtain results within a short period of time. In mining, areas where crushed or broken coal accumulates present the greatest hazard of spontaneous heating.

[4]Pertinent terms are defined in the glossary at the beginning of this report.

Rank.--It is generally agreed that spontaneous combustion is a rank-related phenomenon. As the rank of coal decreases, the hazard of spontaneous heating increases. Lignites and subbituminous coals are most susceptible to spontaneous heating.

Changes in Moisture Content.--The moisture content of the coal, or more exactly, changes in moisture content, affect the tendency of coal to spontaneous heating. Drying, loss of moisture, is an endothermic process and lowers the temperature of the coal; it also exposes more oxidative sites. Wetting, gain of moisture, is an exothermic process, and the heat liberated may be sufficient in itself to cause spontaneous heating. For a given coal, at temperatures below 100° C, the heat of wetting is greater than the heat of oxidation.

Temperature.--The rate of coal oxidation is a direct function of temperature; the higher the temperature, the faster the rate at which coal reacts with oxygen. This is particularly important in areas where heat generated by oxidation accumulates, further accelerating the rate of oxidation. It is also significant in the presence of a thermal anomaly; that is, where the ground temperature is substantially higher than normal.

Pyrite content.--The presence of the sulfur minerals pyrite and marcasite may accelerate spontaneous heating. Under certain conditions, the pyrite may swell and cause the coal to disintegrate, exposing more oxidative sites. If the pyrite is finely divided and can be rapidly converted to ferrous sulfate, the coal is more susceptible to spontaneous heating. Generally, the pyrite concentration must exceed 2 pct before it has a significant effect.

Geological Factors.--The presence of faults in the coal seam may contribute to spontaneous heating by allowing air and water to migrate into the coal seam. Zones of weakness around faults also allow air leakage into the coal mass. When the coal seam under shallow cover is mined, cracks and fissures may develop in the coal and adjacent strata. Air and water from the surface can gain access to the coal and increase the potential for spontaneous heating.

Mining Practice.--Several factors in the mining method, particularly in underground mining, can contribute to potential for spontaneous heating. Areas where fine coal particles accumulate, especially gob areas, present a hazard because of the large coal surface available for oxidation. Air leakage around and through fissured pillars and into abandoned areas of the mine allow coal to oxidize and also allow generated heat to accumulate. Changes in ventilation, either intentional or accidental, may cause air leakage or may suddenly bring moist air into contact with dry coal.

Although all these factors affect spontaneous heating of coal, rank and changes in moisture content are apparently the most important. The degree of importance of the other factors and the relationship among them have not been specifically determined.

INDEX OF COMBUSTIBILITY

A readily determined, universally applicable index of combustibility would be the preferred method of evaluating a coal's liability to spontaneous heating. Although several indices of combustibility are proposed in the literature, none is in general use. The most common indices are described in the following paragraphs.

Heating Rate.--The temperature increase under controlled conditions, measured in degrees Centigrade per hour per gram (° C/hr/g). Another variant involves the measurement of the quantity of heat released, measured in calories per hour per gram (cal/hr/g). The greater the heating rate, the greater is the tendency to spontaneous heating. Two variations of this method are--

> Relative Heating Rate.--The heating rate relative to the heating rate of some standard sample.
>
> Dynamic Heating Rate.--Rate at which the temperature of the coal increases with respect to its surroundings within a given temperature range.

Crossing-Point Temperature.--The temperature at which the temperature of the coal and a heated bath coincide, also called relative ignition temperature.

Ignition Temperature.--The temperature at which the coal ignites. The ignition temperature is determined by observation; that is, the temperature at which an observer sees the coal fire. It cannot be exactly defined and, therefore, can only be used to determine relative spontaneity of the coals tested.

$CO/\Delta O_2$.--The ratio of CO formation to oxygen adsorption, also called the "CO index." The increase in the concentration of CO and the decrease in the concentration of oxygen are related to the temperature of the coal. Monitoring mine air for CO and oxygen is the basis for a method of detecting spontaneous heating.

Combinations of the preceding indices also are used in an attempt to achieve greater accuracy. All of the indices are determined by a laboratory procedure and require standard conditions to obtain reproducible results. In some studies, the index of interest is compared to some internal standard; oxygen content of the coal, for instance, without mathematical correlation. In other studies, a simple ranking system is used to evaluate the coal's combustibility. Indices determined in the laboratory have not been compared to the actual incidence of spontaneous heating. Owing to the lack of an objective criterion and the diversity of experimental conditions under which the indices were developed, comparing various indices would be inaccurate and possibly misleading. No particular index has been shown to be significantly more accurate, and none is in general use. At present, the CO index is probably receiving the most interest in the United States. This is not because of demonstrated superiority as an index of combustibility, but because

of its relatively simple adaptation to the detection of spontaneous heating. In many instances, evaluation of the hazards from spontaneous heating is based on previous experience with a given coal.

EXPERIMENTAL METHODS

The various methods used to study spontaneous heating are described in detail in the literature (22); most are variations of the four basic methods described in the following paragraphs.

Adiabatic Calorimetry.--The coal sample is placed in an insulated container or bath, and the whole system is heated to a preselected temperature. When air or oxygen is added, the temperature of the coal rises; the material surrounding the coal is heated so that its temperature coincides with the measured temperature of the coal. Since there is no heat loss to the surroundings, changes in coal temperature are measured accurately. The change in the temperature of the coal in a given time, the time needed to reach a preselected temperature, or the amount of heat generated per unit of time is used to evaluate the spontaneous heating potential of the coal.

Isothermal Calorimetry.--In this method, a coal sample is placed in a large bath held at a constant temperature. Heat generated in the coal sample, by oxidation or wetting, is measured by thermocouples and dissipated in the relatively large heat sink. The measured rate of heat generation is correlated with the combustibility of the coal.

Oxygen Sorption.--In the O_2-sorption method, a coal sample is placed in a container and air or oxygen is added. In a closed system, gaseous reaction products are periodically removed, and the amount of air or O_2 that must be added to maintain the system at constant pressure is used to estimate the amount of oxygen adsorbed. In a flow system, the flow rate and analysis of the effluent gas are used to estimate the amount of oxygen adsorbed by the coal. The temperature increase per unit of oxygen consumed indicates the coal's potential for spontaneous heating.

Temperature Differential.--The coal sample is placed in a bath and heated at a constant rate; the temperature difference between the coal and the bath is measured. Initially, the temperature of the coal lags behind the bath temperature. When the coal begins to self-heat, the temperature of the coal will coincide with, then exceed, the temperature of the bath. Usually, some variant of a temperature differential-time relationship indicates the combustibility of the coal.

Most experimental studies use one of the four basic methods described, but other factors vary for each experiment. The coal samples vary, particularly in origin, amount, preparation, and particle size. The atmosphere is usually air or O_2, although N_2 or other inert gas may be used for baseline evaluation or to bring the coal to the starting temperature. The oxidizing medium used may be moist or dry, flowing or static. The initial temperature, final temperature, and heating rate also vary. In some cases, the effluent gas is analyzed for CO, CO_2, O_2, CH_4, and other hydrocarbons to provide a more

detailed study of the oxidation rate and reactions. In these methods, the standard of combustibility is internal and relative; that is, if the measured variable is greater for coal A than coal B, coal A is more prone to spontaneous heating than coal B. Although experimental results may be accurate, they cannot be extended to other coals unless the experimental conditions are duplicated exactly.

CONCLUSION

Rank and changes in moisture content are the most important factors affecting the spontaneous heating of coal. Temperature, air flow rate, particle size, and pyrite concentration must also be considered, both in laboratory experiments and in adapting experimental data to mine conditions. In the mine, geological conditions and mining practice affect the spontaneous heating hazard. From information in the literature, it is apparent that two processes, oxidation and wetting, contribute to spontaneous heating. When the low-rank coals of the Western United States are mined, air in contact with the coal, either by diffusion through cracks and fissures in the solid coal or around and through rubble piles, will oxidize the coal, raising its temperature. A difference between the humidity of the air and the moisture content of the coal will cause further temperature changes. Dry air flowing over moist coal will remove moisture and lower the temperature of the coal; moist air in contact with dry coal will cause a temperature increase. The amount of energy released by oxidation and/or wetting of a particular coal will determine the spontaneous heating hazard.

There is no standard method of studying spontaneous heating in the laboratory, but some form of calorimetry is currently most popular. The laboratory method used should be capable of determining the extent of heat generation due to the oxidation, due to wetting, and due to combined oxidation-wetting processes. According to the literature, there are several possible indices of combustibility. However, none is clearly superior or in general use for predicting hazards due to spontaneous heating of coal. At the present time there is no simple, reliable, and objective method of evaluating a coal's potential for spontaneous heating. Past experience, although often inaccurate and unreliable, is the most commonly used indicator of a spontaneous heating hazard.

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APPENDIX.--SUMMARY OF IMPORTANT PAPERS

	Berkowitz (5)	Bhattacharyya (6)	Bhattacharyya (7)	Carpenter (9)	Elder (16)	Feng (17)
Coal studied:						
Country of origin	Australia	Great Britain	Great Britain	Wales	United States	Canada.
Fixed carbon	Lignite-bituminous	67 to 95 pct maf	67 to 95 pct maf	87 to 95 pct maf	50 to 81 pct maf	70 to 87 pct maf
Sample condition:						
Moisture	Dried	Variable	Variable	Dried	Dried	NR
Particle size	Minus 40 BSS[1]	Minus 72 BSS[1]	Minus 72 BSS[1]	72/100 BSS[1]	1/4 inch	NR
Sample size....g	40	5	5	20	9,000	NR
Atmosphere:						
Content	Moist O_2	Air or N_2	Air or N_2	Dry O_2	O_2	NR
Flow rate	1,980 ml/min	2.5 ml/min/g	2.5 ml/min/g	0	Variable	NR
Temperature....° C	25, 40	30	30	75 to 115	27 to 150	NR
Method	Adiabatic calorimetry.	Isothermal calorimetry.	Isothermal calorimetry.	O_2 sorption	Adiabatic calorimetry.	Temperature differential.
Combustibility index	NA_p	Heating rate	Heating rate	NA_p	Relative heating rate.	Heating rate divided by ignition temperature.
Results	Heat of wetting causes greater temperature increase than heat of oxidation.	Sorption of H_2O is rate-determining step. Rate of heat release increases with increased difference in humidity of coal and atmosphere.	Desorption of H_2O results in heat loss. Rate of heat loss increases with difference in humidity of coal and air.	Chemisorption is rate-determining step for oxidation. Rate of O_2 adsorption controlled by pore diameter.	Relative heating rate is an exponential function of O_2 concentration.	Hazard of spontaneous combustion is a combination of coal properties and mine environment.

See footnotes at end of table.

	Güney (22)	Güney (19)	Güney (23)	Hodges (26)	Hodges (24)	Nandy (32)
Coal studied:						
Country of origin.	NR	Great Britain..	Great Britain..	Great Britain...	Great Gritain...	India..........
Fixed carbon......	NR	81 to 86 pct maf.	59 to 64 pct maf	81 to 84 pct maf	60 to 65 pct maf	46 to 78 pct maf
Sample condition:						
Moisture.........	Dried	Dried	Dried	Dried and as-received	Dried or moist..	Moist..........
Particle size.....	Minus 72 BSS[1]	Minus 0.21 mm	Minus 72 BSS[1]	Minus 72 BSS[1]	Minus 72 BSS[1]	Minus 72 BSS[1]
Sample size....g..	100	100	100	2,000	4 to 5	20
Atmosphere:						
Content..........	Air, moist or dry.	Moist air......	Moist air......	O_2, dry or moist.	Air............	Moist air.......
Flow rate........	Variable	250 ml/min	250 ml/min	1,500 ml/min	10 ml/min	80 ml/min
Temperature....° C..	40 to 60	40	40	30	30	75
Method............	Adiabatic calorimetry.	Adiabatic calorimetry.	Adiabatic calorimetry.	Adiabatic calorimetry.	Adiabatic calorimetry.	Temperature differential.
Combustibility index	NA_p	Heating rate...	Heating rate...	NA_p	NA_p	Crossing-point temperature.
Results...........	In adiabatic calorimeter, greatest temperature increase occurred with dry coal and moist air.	Greatest temperature increase related to oxidation and H_2O adsorption. Difference in humidity of air and coal significantly influences reaction rate.	ΔT:Oxidation, <4° C; dry coal, moist air, 10°-25° C; moist coal, dry air, -3° C. CO index did not correspond with empirical evaluation of susceptibility to spontaneous heating.	Moisture accelerates heating of dry coal. Less heating occurs with moist coal.	Heat generated is 2.5 times greater for moist oxidation than for dry oxidation. ΔH: Oxidation, <5 cal/g; evaporation, -10 cal/g; and wetting, >30 cal/g.	Higher incidence of spontaneous combustion with high-moisture coals. Low crossing-point temperature correlated with high susceptibility to spontaneous combustion.

See footnotes at end of table.

	Oreshko (33)	Panaseiko (34)	Sevenster (36)	Sevenster (37)	Stott (40)
Coal studied:					
Country of origin	U.S.S.R.	U.S.S.R.	South Africa	South Africa	NR
Fixed carbon	NR	NR	62 to 71 pct maf	62 to 71 pct maf	52.6 pct maf.
Sample condition:					
Moisture	NR	Dried, moist	Dried	Dried	Dried.
Particle size	NR	NR	Minus 60 BSS[1]	Minus 60 BSS[1]	1/10 inch to minus 60 BSS[1].
Sample size....g	NR	NR	5	5	3,000
Atmosphere:					
Content	NR	O_2	O_2	O_2	Moist O_2, air
Flow rate	NR	0	0	0	1,000 ml/min
Temperature....° C	NR	20	-180 to 100	30 to 40	22
Method	Temperature differential.	O_2 sorption	O_2 sorption	O_2 sorption	Isothermal calorimetry.
Combustibility index	Ignition temperature.	NA_p	NA_p	NA_p	NA_p
Results	Plot of weight of coal versus temperature shows maximum weight at transition between controlled oxidation and burning.	Moisture either accelerates or retards oxidation depending upon state of H_2O and degree of metamorphism of coal. A critical moisture content varies with rank of coal.	Between 0° and 100° C, adsorption increases with temperature; chemisorption and reaction occur above 40°. Coal-O_2 complex forms above 72°. Rate of O_2 adsorption decreases with time.	Initial O_2 adsorption is chemisorption; accounts for 5 pct of adsorption. Adsorption influenced by diffusion rate and chemical reaction.	Heat of wetting greater than heat of oxidation. Air or O_2, saturated, caused spontaneous ignition of dried coal.

NA_p--Not applicable; NR--Not reported.
[1] British sieve size.

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